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(54) Title: PLASTIC FILMS AND ROLLS FOR II	N-MOI	D LABELING, LABELS MADE BY PRINTING THEREON,

(54) Title: PLASTIC FILMS AND ROLLS FOR IN-MOLD LABELING, LABELS MADE BY PRINTING THEREON,
AND BLOW MOLDED ARTICLES LABELED THEREWITH

#### (57) Abstract

Polymeric sheets or rolls suitable for printing and forming in-mold labels for plastic containers comprise a polymeric transparent, translucent or contact clear substrate having a thickness in the range of 0.002 to 0.008 inches which is coated on the container-facing side with a heat activatable adhesive. Such a sheet or roll can be printed with label indicia on the exposed face or reverse printed on the face covered by the adhesive and then cut into individual labels for affixing to the container as part of the in-molding process. Recyclable containers are provided having firmly adherent, squeeze-release resistant labels and the indicia in preferred embodiments are protected from spillage because they are viewable through the labels themselves.

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# PLASTIC FILMS AND ROLLS FOR IN-MOLD LABELING, LABELS MADE BY PRINTING THEREON, AND BLOW MOLDED ARTICLES LABELED THEREWITH

FIELD OF THE INVENTION

This invention relates to polymeric sheets or rolls particularly adapted for use in the in-mold labeling of blow-molded plastic containers. More particularly, the present invention relates to transparent, translucent or contact clear films having a heat activatable adhesive thereon and adapted for printing and use as labels in such in-mold labeling applications.

BACKGROUND OF THE INVENTION

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Plastic containers or bottles are prevalent today in a wide variety of shapes and sizes for holding many different kinds of materials such as light duty liquids (e.g., dishwashing detergent), heavy duty liquids (e.g., laundry detergents), motor oil, vegetable oil, herbicides, etc. Generally, these containers are fabricated from layers or a plurality of layers of plastic, particularly polypropylene, polyethylene and polyesters, particularly poly(ethylene terephthalate), by means of blow molding or injection molding.

Generally such containers are provided with a label which designates the trade name of the product and may contain other information as well. In some instances, the label is merely attached to the container after molding by means of adhesive or the like. However, the label may also be attached to the container during the container molding process. This technology by which the label is associated with the container during the molding operation is generally referred to as an in-mold label process.

Many devices are known for performing in-mold labeling of a plastic container. For example, German Published Pat nt Application No. 1,807,766 to Rosler et al in 1969 shows making plastic containers label d by in-mold t chniques with a transparent plastic film rev rse-printed

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with indicia viewable through the film. There is no adhesive layer between the label and the hollow plastic container, however, and the bottle label relies on the attraction between the polymeric label film and the plastic container body for adhesion. This means of adhesion, particularly in squeezable containers, has been a continuing problem, because such labels will not remain adhered to the container but instead they will split or separate because the finished container is normally bent, flexed and squeezed during use.

In U.S. Patent 4,837,075 issued to Dudley in 1989, there is shown a coextruded plastic film label for in-mold labeling comprising a heat activatable ethylene copolymer adhesive layer and a surface printable layer comprising polystyrene with optional intermediate layers to provide interlayer adhesion and recycle of reground labels. The label has preferably a thickness in the range of from about 0.002 to 0.005 inches (0.05 to 0.127 mm). Because the layers have different refractive indices and to hide blemishes, the patent (Col.3, lines 46-60) teaches the need to add pigment or fillers to provide a suitable background for printing. This has the effect of making the label opaque and it will no longer be suitable for example to reverse print the back side of the label and thereafter view the printed indicia therethrough.

In published European Patent Application No. 0,281,701 issued to Court et al in 1988, there is disclosed an in-mold label formed from a thin sheet of multicellular thermoplastic film composed of a biaxially extruded opaque, non-transparent polyethylene/polystyrene copolymer for use with a blow molded plastic container. The in-mold label is said to resist curling, wrinkling and crazing, to have thermodynamic pr perties similar t those in the plastic of the container, and may b r cycled along with the container. Such multicellular films do not, however, have the sam

specific gravity as the plastic from which they are made because they are filled with gas-containing Furthermore, because the films are not transparent, translucent or even contact clear they must be printed on the outside and then overcoated with a surface coating to insure that the printed indicia does not smear or rub off (Page 4, lines 46-47). Printing is suggested on the inner surface of the backing (Page 5, lines 16-18), but only if the label is to be used as a peel-off label apparently because in such event the indicia cannot be read through the opaque label material. Such labels have the further disadvantage that they cannot be applied to containers and then reground in high volume after use to make recycled materials for blow molding containers because the container colors will not match the same color printed on the label.

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The present state of the art is further evidenced by Modern Plastics, September, 1990, in an article relating to plastics used for labels, pages 83-85. It is stated therein that labelstock producers are still going after inmold labeling with a variety of products. Among the materials under active investigation was mentioned paperlike polyolefin films. However, work has shown that such films are only commonly manufactured in thicknesses of up to about 0.0017 mil and they therefore are not thick enough to provide the necessary strength to withstand the heat of the molding process, even if they are reverse printed to resist staining from spillage and even though such materials assist in recycling. They are also too thin to pick and place in the mold.

It has now been discovered that transparent, translucent, clear or contact clear polymeric films having judiciously s lected characteristics of thickness, specific gravity and coefficient of expansion and contraction and provided with a heat activatable adh sive coating hav

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improved and surprising characteristics of adhesion to in-mold blown plastic containers with resistance to damage from cracking, tearing, creasing, wrinkling or shrinking due to physical abuse and flexing of the plastic container material. Furthermore, if such sheets or rolls are reverse printed on the back before being overcoated with the adhesive, and labels made therefrom are then affixed to bottles during the blow molding process, abrasion scuffing and product spillage will not adversely affect the function of the label. Optionally, if a metallized coating of a thin metal film is deposited on the sheets or rolls, premium quality, decorative labels with all of the advantages set forth above will be provided.

Accordingly, a principal object of the present invention is to provide printable polymeric sheets or rolls for labels for in-mold use without the problems discussed above. It is a further object of the invention to provide a method for in-mold labeling of hollow plastic containers using printed labels made from such sheets. It is still another object of the invention to provide articles labeled with printed labels which have the unexpectedly superior properties described above.

These and other objects of the invention will become apparent from the present specification.

### SUMMARY OF THE INVENTION

According to this invention, in one of its major aspects, there are provided polymeric sheets or rolls particularly adapted for use in the in-mold labeling of blow-molded plastic containers, the sheets or rolls comprising:

(1) a layer of a transparent, translucent or contact clear substrate formed from a polymer or a mixture of polymers, the layer being surface printable or capable of being rendered surface printabl, having a thickness between 0.002 and 0.008 inches, having a specific gravity substantially the same as that of the polymer or mixture of

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polymers from which it is formed, and having a coefficient of thermal expansion or contraction under the conditions which the container sees the same or substantially the same as that of the plastic from which said container is made;

- (2) a print-receiving area on one major face of layer (1); and
- (3) a heat activatable adhesive layer covering all or a patterned part of an exposed major face of layer (1) or the print-receiving area (2), the polymeric sheet or roll being regrind compatable with the plastic containers which are to be blow molded around labels made from it, the adhesive being characterized by the ability to form a bond with the substrate and the plastic container such that the strength of the substrate-adhesive interface and the plastic containeradhesive interface and the cohesive strength of the adhesive itself are all greater than the forces required for deformation and recovery of the film itself, whereby labels made from the polymeric sheet have improved and surprising characteristics of adhesion to the plastic container with resistance to damage from cracking, tearing, creasing, wrinkling or shrinking due to physical abuse and flexing of the plastic container material.

In preferred features, the invention contemplates a polymeric sheet or roll as defined above which also includes

(4) a thin metal coating on at least one of the major faces of substrate layer (1), especially a sheet wherein the thin metal coating (4) comprises a vacuum metallized coating of aluminum. Also provided are such sheets or rolls wherein the layer of substrate (1) comprises a clear or contact clear virgin, recycled or reprocessed polyester; a polymeric sheet or roll as defined above wherein the layer of substrat (1) comprises a clear or contact clear virgin, recycled or reproc ssed olefin homopolymer or copolymer or a blend th reof; a polymeric sheet or roll as defined above

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further comprising printed indicia on said print-receiving area (2) of the substrate, or on the substrate at the interface with said adhesive layer (3), or at both locations; especially such a polymeric sheet or roll wherein the indicia are reverse printed on the substrate at the interface with the adhesive layer (3) and the adhesive layer overcoats the sheet completely or in a defined pattern whereby after affixing labels made from the sheet or roll to a plastic container during the blow molding process there will be captured a printed image equivalent to a front label between the label substrate and the container thus protecting the label from the contents of said container when spilled, from the environment and from abrasion when the container is processed, shipped or used. Particularly preferred are polymeric sheets or rolls wherein the printing is carried out in three stages so as to produce a first indicia which is reverse printed, a second overprinted indicia which is opaque and a third overprinted indicia which is direct printed whereby after affixing labels made from such sheets or rolls to a clear or translucent plastic container during the blow molding process there will be provided in one label the equivalent of a front and back label when the container is empty or partially or completely filled with a clear or translucent liquid. Also contemplated is a polymeric sheet or roll as above defined wherein said printed indicia is provided with a primer for sealing the printed image and to enhance adhesive bonding if desired; a polymeric sheet or roll as first above defined wherein the heat activated adhesive layer (3) is a coated layer; a polymeric sheet or roll as defined above wherein the heat activated adhesive layer (3) comprises an ethylene/vinyl acetate copolymer. The invention also provides a polymeric sheet or roll as defined above wher in the print-receiving face of the substrate (1) has been treated to enhance ink

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anchorage by application of a primer, by flame treatment, by corona-treatment or by chemical treatment.

In another major aspect, the invention provides in-mold labels for plastic containers, the labels comprising:

- (1) a layer of a transparent, translucent or contact clear substrate formed from a polymer or a mixture of polymers, the layer being surface printable or capable of being rendered surface printable, having a thickness between 0.002 and 0.008 inches, having a specific gravity substantially the same as that of the polymer or mixture of polymers from which it is formed, and having a coefficient of thermal expansion or contraction under the conditions which the container sees the same or substantially the same as that of the plastic from which said container is made;
- (2) a print-receiving area on one major face of layer (1); and
- or a patterned part of an exposed major face of layer (1) or the print-receiving area (2), the label being regrind compatable with the plastic containers which are to be blow molded around labels made from it, the adhesive being characterized by the ability to form a bond with the substrate and the plastic container such that the strength of the substrate-adhesive interface and the plastic container-adhesive interface and the plastic container-adhesive interface and the forces required for deformation and recovery of the film itself, the label further comprising printed indicia on the print-receiving area (2) of the substrate, or on the substrate at the interface with the adhesive layer (3), or at both locations.

Still another major aspect of the invention provides plastic contain rs, esp cially thos compris d f single or multilayers of polypropyl ne, polyethylene or polyester, of the typ produc d by blow molding, including improved in-mold

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labels adhered to the outer surface of the containers, the labels comprising:

- a layer of a transparent, translucent or (1) contact clear substrate formed from a polymer or a mixture of polymers, the layer being surface printable or capable of being rendered surface printable, having a thickness between having a specific 0.002 and 0.008 inches, substantially the same as that of the polymer or mixture of polymers from which it is formed, and having a coefficient of thermal expansion or contraction under the conditions which the container sees the same or substantially the same as that of the plastic from which said container is made;
- (2) a print-receiving area on one major face of layer (1); and
- or a patterned part of an exposed major face of layer (1) or the print-receiving area (2), the label being regrind compatable with the plastic containers which are to be blow molded around labels made from it, the adhesive being characterized by the ability to form a bond with the substrate and the plastic container such that the strength of the substrate-adhesive interface and the plastic container-adhesive interface and the forces required for deformation and recovery of the film itself, the label further comprising printed indicia on said print-receiving area (2) of the substrate, or on the substrate at the interface with said adhesive layer (3), or at both locations.

In a further major aspect, the present invention contemplates: a method of producing an in-mold plastic container comprising:

- (a) providing a polymeric she t comprising:
- (1) a layer of a transparent, translucent or contact clear substrate formed from a p lymer or a mixture

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of polymers, the layer being surface printable or capable of being rendered surface printable, having a thickness between 0.002 and 0.008 inches, having a specific gravity substantially the same as that of the polymer or mixture of polymers from which it is formed, and having a coefficient of thermal expansion or contraction under the conditions which the container sees the same or substantially the same as that of the plastic from which said container is made;

- (2) a print-receiving area on one major face
  10 of layer (1); and
  - all or a patterned part of an exposed major face of layer (1) or the print-receiving area (2), the polymeric sheet or roll being regrind compatable with the plastic containers which are to be blow molded around labels made from it, the adhesive being characterized by the ability to form a bond with the substrate and the plastic container such that the strength of the substrate-adhesive interface and the plastic container-adhesive interface and the cohesive strength of the adhesive itself are all greater than the forces required for deformation and recovery of the film itself;
  - (b) printing indicia on the print-receiving area(2) of the substrate, or on the substrate at the interface with the adhesive layer (3), or at both locations;
  - (c) cutting said sheet into a plurality of individual labels, each label bearing printed indicia;
    - (d) positioning one of the labels in a mold;
  - (e) blow molding a plastic container, preferably comprised of a single or a multilayer of polypropylene, polyethylene or polyester, within the mold such that the label adheres to the outer surface of the blow molded container; and
    - (f) removing the container from the mold.

## DETAILED DESCRIPTION OF THE INVENTION

The terms "virgin", "recycled" or "reprocessed" when used herein and in the appended claims mean, respectively, new resin, reground resin, and resin sheets and the like which have been prepared for other uses, and after-treated to remove coatings, etc.

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The term "regrind compatible" when used herein and in the appended claims means that containers with in-mold labels can be reground and molded after being mixed with virgin material. Regrind compatibility is determined by regrinding, mixing and molding.

The term "contact clear" when used herein and in the appended claims means a hazy material difficult to see through, but which, in intimate contact with a surface, transmits an underlying image. Polyethylene films are a common example. Contact clarity is determined by a simple trial and error test.

The terms "primer", "flame-treatment", "corona treatment", and "chemical treatment" when used herein and in the appended claims mean, respectively, a deposited coating for promoting adhesion generally comprising a filled or unfilled polymer, surface activation by carefully exposing to a bank of flames, without burning or distortion, exposure to high voltage direct current to microscopically etch the surface, and carefully etching the surface with chemicals known to be effective for this purpose.

The labels of the invention comprise a substrate which has characteristics substantially similar to the plastic container with which the label is to be used with special reference to the polymers used. This prevents loosening of the label, especially at its edg s after the in-mold processing and facilitat s recycling.

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The substrate film can or cannot be oriented, depending on how it is made. As is well known, cast film can or cannot be oriented, but is usually oriented to a minor degree in the machine direction (MD). Blown film is usually oriented due to the manufacturing process, but is not usually sold as oriented because it is an unbalanced orientation. Extruded film is usually oriented to a major degree, and orientation can be monoaxial or biaxial. Although any such film can be used in the present invention, it is preferred to use monoaxially or biaxially oriented film as the substrate.

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The substrate should have a "specific gravity substantially the same as that of the polymer or mixture of polymers from which it is formed." This gives the labels sufficient strength and facilitates their use in label transfer devices in automated molding equipment. The specific gravity is measured in standard ways, e.g., by American Society of Testing Materials (ASTM) Standard Test Method D782 and, if the values are reported in units of g/cm, variations of plus or minus 15 percent as between label and the source polymer are permissible as being within the meaning of "substantially the same." The substrate should have "a coefficient of thermal expansion or contraction under the conditions which the container sees the same or substantially the same as that of the plastic from which said container is made." Some variability is permissible, and characteristic seems to be a factor in preventing lifting of the edges of the blow molded containers bearing the in-mold labels of the invention. Coefficient of thermal expansion or contraction is measured by standard methods, such as by ASTM Method D696, which expresses the values in units of 10-6 in/in/°C, or in values of % / °C from which the permissible variations m ntioned her inabove are m asured. How ver, th best test is a practical one: make a t st contain r and subject it to a heat and cooling cycling in a controlled

temperature oven. Those combinations of label materials and bottle plastics free of edge lifting are suitable.

A heat activated adhesive is applied to a surface of the substrate which will come into contact with the container. Selected inkwork comprising printed indicia is printed on the opposite surface of the substrate or, in preferred embodiments the indicia will be reverse printed on the back surface, i.e., under the adhesive coating or, if desired, on both surfaces. The label is then positioned in a blow mold prior to container formation. As the container is formed, the adhesive is activated by the heat of the mold and its contents and adheres the label to an outer surface of the container.

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The preferred embodiments of the labels of the present invention are fabricated from transparent, translucent or contact clear extruded, cast or blown films of polyolefin, e.q., polyethylene or polypropylene or polyester and these may optionally be provided with a print enhancing coating such as those well known to those skilled in this art. The films are provided in sheets or rolls which may be printed with conventional label indicia on conventional printing equipment and furthermore can be die cut and applied to plastic containers using conventional in-mold equipment. Although for purposes of exemplary showing, the present invention is described and illustrated in connection with a polyethylene container, it will be understood that in-mold labeling may also be applied in the formation of propylene multi-layer bottles, polyethylene terephthalate bottles and other types of plastic containers formed by blow molding.

The preferred construction of the improved in-mold labels of the present invention uses a solid, i.e., non-multicellular thermoplastic film comprised f a polypropylene polymer in a biaxially extruded single ply. Such films are marketed und r the name "TREAX<sup>®</sup>" by Toray Industries, North

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Kingstown, RI, 02852, U.S.A. Other preferred films are virgin and reprocessed poly(ethylene terephthalate) film, such as those marketed under the name "MYLAR®" and "REVVAL", respectively, by DuPont, Wilmington, DE, U.S.A. In order to enhance the printing qualities of the thermoplastic film it may be provided with, for example, a filled, e.g., filled with clay/calcium carbonate, silica and/or china clay, etc., or an unfilled primer coating, such as a saran type resin. Typically such primers are available comercially from sources well known to those skilled in this art. For example, polyester and acrylic primers are marketed under the name "ADCOTE" by Morton International, Inc., Chicago, IL, U.S.A. The coating helps insure that the surface of the film will accept high quality printing and may also improve the abrasion and scuff resistant qualities of the finished label.

The physical properties of the aforementioned optically clear, biaxially oriented thermoplastic polypropylene film (TREAX® 300), are set forth in Table 1:

	TABLE 1	
20	Density Thickness	0.905 g/cm <sup>3</sup> 0.003 inches
	Tensile Strength, MD TD	16,000 psi 28,000 psi
25	Elongation at Break, MD TD	160% 60%
	Folding Endurance Coefficient of Expansion**	Excellent 81-100 X 10 <sup>-6</sup> in/in/°C
30	<pre>% Shrink at 275° F, MD,TD Surface treatment</pre>	< 2% Corona-discharge

<sup>--</sup>MD = machine direction; TD = transverse direction
\*\*--Modern Plastics Encyclopedia, October 1989, page 606

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The physical properties of the aforementioned reprocessed poly(ethylene terephahalate) ("REVVAL $^{\oplus}$ ") film are as set forth in Table 2:

5	TABLE 2	TABLE 2				
10	Density Thickness Tensile Strength Elongation at Break Initial Tearing Resistance Young's Modulus(@ 1%)	1.388-1.395 g/cm <sup>3</sup> 0.007 inches 15 kN/cm 70% 3.5 kN/cm <sub>2</sub> thickness 370 kN/cm <sup>2</sup>				
15	Folding Endurance Coefficient of Expansion % Shrink at 275° F, MD,TD Surface treatment	over 15,000 cycles About 0.002 %/°C < 0.2% Saran resin coating				

--MD = machine direction; TD = transverse direction

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A heat activated adhesive is applied to such label sheets in a conventional manner. The use of such coatings for in-mold labels is reviewed in detail by D.H. Wiesman in Tappi Journal, Vol 69, No. 6, June 1986. A preferred adhesive comprises an organic polymeric resin such as an ethylene/vinyl acetate copolymer gel or dispersion. A suitable source of such adhesives is Morton International Corp. which sells such products under the name "ADCOTE®" 31DW1974 (Solvent-based) and X19-23 (Water-based). Also suitable is a warm melt adhesive designated Product No. S11723 and sold by Selective Coatings & Inks, Inc., Farmingdale, NJ, U.S.A. Before, or after (if reverse printing is employed), applying the adhesive, the film is printed with suitable label indicia in a conventional manner. The adhesive is preferably applied so as to produce a smooth, continuous coating. Although it can also be applied by a gravure coating process, this may produce a patterned visual image, which is not desirable on clear blow-molded bottles. It has been found that the printing quality of the present thermoplastic film labels is quival nt to the printing quality of conventional pap r labels. Finally,

individual labels may be die cut from the sheets or rolls in the conventional manner.

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With respect to printing, various methods are used in this art to apply information or decorations to plastics, Indeed, most of the processes for printing on paper also apply to printing on plastics. The most common printing techniques used are gravure and flexographic for layflat film products, screen or jet printing, and pad transfer and web printing for traditional printing on labels. To avoid unnecessarily detailed description, reference is made to Modern Plastics Encyclopedia, Mid-October Issue, 1989, "Printing" by Hans Deamer, pages 381-383.

Selection of the printing inks for use, and formation of print-enhancing surfaces and the production of images or indicia are well within the skill of workers in this field. Also, it is easily obvious to the artisan to produce the films of this invention with printed and reverse printed indicia on any print-receiving surface and to carry out the printing operation in the stages set forth in the description above. The inclusion of primers for sealing the printed image and to enhance adhesive bonding is also conventional in this art.

For those embodiments requiring them, metallized coatings are also conventionally applied. For example, a thin coat of aluminum can be applied to one surface of the film which may already have been printed either directly, in reverse, or both directly and in reverse. The metal film enhances customer acceptance in some products, especially premium products. For example, reverse printing achieves an underglaze three-dimensional effect that is enhanced by the high gloss imparted by the mirror-like metallized coating. A preferr d process of applying a thin coat of metal is known as vacuum metallizing. In such a proc ss a material, e.g., aluminum, is evaporated int a vacuum and a thin metal coating

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is deposited on the substrate. Coating thicknesses are generally of the order of 400 to 500 Angstroms. Vacuum metallizing is a so-called line-of-sight process, meaning that the coating is applied only on a surface that is exposed to the source. Vacuum metallizers are either batch or continuous coaters. A detailed description of such metallizers and their use to coat plastic films and rolls is provided in Modern Plastics Encyclopedia, <u>Ibid.</u>, "Vacuum Metallizing" by D.A. Lombardo and L.E. McCrary, pages 384-385. It is preferred to use herein a conventional batch metallizer of the web metallizing type. A roll of plastic film will be the substrate and the evaporant is preferably aluminum evaporated diboride-boron nitride, or the titanium intermetallic composite boats.

The in-mold labels of the present invention may be utilized on conventional in-mold labeling apparatus in the same manner as conventional paper labels. See, for example, the article in Tappi Journal, cited above.

To save unnecessarily detailed description, devices for performing in-mold labeling on a container, which are well known, are the subject matters of U.S. Patent No. 3,759,643 to Langecker, 1973, and 4,479,644 to Bartlmee et al, 1984. In general, all such apparatus use a blow mold having a cavity for containing a hollow body, and a member which is movable toward the cavity. The member includes a section for carrying a label to be applied the the hollow body during movement of the member toward the cavity. Ventilation openings are provided in the mold for venting any air between the hollow body and label. Variations in the apparatus that may be employed include using rotating mold units and oscillating means for picking up individual labels and depositing them in the rotating molds at appropriate intervals to automate the process.

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The patents, applications, publications and test methods mentioned above are incorporated herein by reference.

Many variations of the present invention will suggest themselves to those skilled in the art in light of the above detailed description. For example, instead of virgin oriented polypropylene as the face film, virgin poly(ethylene terephthalate), polyamide, polyethylene, polycarbonate, fluoropolymers and polyimide films can be used. Instead of 0.007 inch polyester film, 0.004 inch polyester film can be used. Instead of ethylene/vinyl acetate as the heat activated adhesive layer, low density polyethylene can be used. Instead of an acrylic printing enhancing coating, another coating, such as a polyester or urethane resin containing finely divided clay or silica, can be spread on the print receiving face of the polymeric sheet or roll. Instead of a polyethylene container, a polypropylene container or a polyester container, the labels can be applied to containers made by blow molding single or multi-layers of barex, cellulose acetate, cellulose acetate butyrate, cellulose acetate propionate, ionomer resin, K-resin, polystyrene and poly(vinyl chloride). Polypropylene labels can be put on polyethylene containers and polyethylene labels can be put on polypropylene containers.

Instead of a single layer label, the labels can comprise two, or more, layers. For example, the outer layer can comprise a reverse printed transparent substrate, comprising at least one layer, through which the indicia is viewed, and this can be laminated to at least one second film, which can be clear or white, for example, and which is(are) unprinted, but which bear(s) the heat activatable adhesive layer, the second film(s) having all of the thermodynamic characterictics required for the first, and serving to anchor the composite label to the blow molded container.

All such obvious modifications are within th full int nded scope of th appended claims.

#### CLAIMS:

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- 1. A polymeric sheet or roll particularly adapted for use in the in-mold labeling of blow-molded plastic containers, said sheet or roll comprising:
- (1) a layer of a transparent, translucent or contact clear substrate formed from a polymer or a mixture of polymers, said layer being surface printable or capable of being rendered surface printable, having a thickness between 0.002 and 0.008 inches, having a specific gravity substantially the same as that of the polymer or mixture of polymers from which it is formed and having a coefficient of thermal expansion or contraction under the conditions which the container sees the same or substantially the same as that of the plastic from which said container is made;
- (2) a print-receiving area on one major face of layer (1); and
  - (3) a heat activatable adhesive layer covering all or a patterned part of an exposed major face of layer (1) or said print-receiving area (2), said polymeric sheet or roll being regrind compatable with the plastic containers which are to be blow molded around labels made from it, said adhesive being characterized by the ability to form a bond with said substrate and said plastic container such that the strength of the substrate-adhesive interface and the plastic containeradhesive interface and the cohesive strength of the adhesive itself are all greater than the forces required for deformation and recovery of the film itself, whereby labels made from said polymeric sheet or roll have improved and surprising characteristics of adhesion to the plastic container with resistance to damage from cracking, tearing, creasing, wrinkling r shrinking due to physical abuse and flexing f the plastic container material.

- 2. A polymeric sheet or roll as defined in Claim 1 which also includes
- (4) a thin metal coating on at least one of the major faces of substrate layer (1).
- 3. A polymeric sheet or roll as defined in Claim 2 wherein said thin metal coating (4) comprises a vacuum metallized coating of aluminum.
- 4. A polymeric sheet or roll as defined in Claim 1 wherein said layer of substrate (1) comprises a clear or contact clear virgin, recycled or reprocessed polyester.
- 5. A polymeric sheet or roll as defined in Claim 1 wherein said layer of substrate (1) comprises a clear or contact clear virgin, recycled or reprocessed olefin homopolymer or copolymer or a blend thereof.
- 6. A polymeric sheet or roll as defined in Claim 1 further comprising printed indicia on said print-receiving area (2) of said substrate, or on said substrate at the interface with said adhesive layer (3), or at both locations.
- 7. A polymeric sheet or roll as defined in Claim 6 wherein said indicia are reverse printed on said substrate at the interface with said adhesive layer (3) and said adhesive layer overcoats the sheet or roll completely or in a defined pattern whereby after affixing labels made from said sheet or roll to a plastic container during the blow molding process there will be captured a printed image equivalent to a front label between the label substrate and the container thus protecting said label from the contents of said container when spilled, from the environment and from abrasion when the container is processed, shipped or used.

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8. A polymeric sheet or roll as defined in Claim 7 wherein said printing is carried out in three stages so as to produce a first indicia which is reverse printed, a second overprinted indicia which is opaque and a third overprinted indicia which is direct printed whereby after affixing labels made from said sheet or roll to a clear or translucent plastic container during the blow molding process there will be provided in one label the equivalent of a front and back label when the container is empty or partially or completely filled with a clear or translucent liquid.

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- 9. A polymeric sheet or roll as defined in Claim 7 wherein said printed indicia is provided with a primer for sealing the printed image and to enhance adhesive bonding if desired.
- 10. A polymeric sheet or roll as defined in Claim
  1 wherein said heat activated adhesive layer (3) is an coated
  layer.
- 11. A polymeric sheet or roll as defined in Claim 1 wherein said heat activated adhesive layer (3) comprises an ethylene/vinyl acetate copolymer.
- 12. A polymeric sheet or roll as defined in Claim 1 wherein the print-receiving face of said substrate (1) has been treated to enhance ink anchorage by application of a primer, by flame-treatment, by corona-treatment or by chemical treatment.
- 13. A flexible and deformable label for deformable plastic containers, said label comprising:
- (1) a layer of a transparent, translucent or contact clear substrate formed from a polymer or a mixture of polymers, said layer being surface printable or capable of being rendered surface printabl, having a thickness between 0.002 and 0.008 inches, having a specific gravity substantially the same as that of the polymer r mixture of

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polymers from which it is formed, and having a coefficient of thermal expansion or contraction under the conditions which the container sees the same or substantially the same as that of the plastic from which said container is made;

- (2) a print-receiving area on one major face of layer (1); and
- or a patterned part of an exposed major face of layer (1) or said print-receiving area (2), said polymeric sheet being regrind compatable with the plastic containers which are to be blow molded around labels made from it, said adhesive being characterized by the ability to form a bond with said substrate and said plastic container such that the strength of the the substrate-adhesive interface and the plastic container-adhesive interface and the cohesive strength of the adhesive itself are all greater than the forces required for deformation and recovery of the film itself, said label further comprising printed indicia on said print-receiving area (2) of said substrate, or on said substrate at the interface with said adhesive layer (3), or at both locations.
  - 14. A label as defined in Claim 13 which also includes
  - (4) a thin metal coating on at least one of the major faces of substrate layer (1).
  - 15. A label defined in Claim 14 wherein said thin metal coating (4) comprises a vacuum metallized coating of aluminum.
  - 16. A label as defined in Claim 13 wherein said layer of substrate (1) comprises a clear or contact clear virgin, recycled or reprocessed polyester.

- 17. A label as defined in Claim 13 wherein said layer of substrate (1) comprises a clear or contact clear virgin, recycled or reprocessed olefin homopolymer or copolymer or a blend thereof.
- 18. A label as defined in Claim 13 further comprising printed indicia on said print-receiving area (2) of said substrate, or on said substrate at the interface with said adhesive layer (3), or at both locations.
- 19. A label defined in Claim 18 wherein said indicia are reverse printed on said substrate at the interface with said adhesive layer (3) and said adhesive layer overcoats the sheet completely or in a defined pattern whereby after affixing said label to a plastic container during the blow molding process there will be captured a printed image equivalent to a front label between the label substrate and the container thus protecting said label from the contents of said container when spilled, from the environment and from abrasion when the container is processed, shipped or used.

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- 20. A label as defined in Claim 19 wherein said printing is carried out in three stages so as to produce a first indicia which is reverse printed, a second overprinted indicia which is opaque and a third overprinted indicia which is direct printed whereby after affixing said label to a clear or translucent plastic container during the blow molding process there will be provided in one label the equivalent of a front and back label when the container is empty or partially or completely filled with a clear or translucent liquid.
- 21. A label as defined in Claim 13 wherein said printed indicia is provided with a primer for sealing the printed image and to enhanc adh sive bonding if desired.
- 22. A label as defined in Claim 13 wh r in said heat activated adh sive layer (3) is a coated lay r.

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- 23. A label as defined in Claim 13 wherein said heat activated adhesive layer (3) comprises ethylene/vinyl acetate copolymer.
- 24. A label as defined in Claim 13 wherein the print-receiving face of said substrate (1) has been treated to enhance ink anchorage by application of a primer, by flametreatment, by corona-treatment or by chemical treatment.
- 25. In a plastic container of the type produced by blow molding, the improvement in combination therewith comprising an in-mold label adhered to the outer surface of said container, said label comprising:
- (1) a layer of a transparent, translucent or contact clear substrate formed from a polymer or a mixture of polymers, said layer being surface printable or capable of being rendered surface printable, having a thickness between and 0.008 inches, having a specific substantially the same as that of the polymer or mixture of polymers from which it is formed, and having a coefficient of thermal expansion or contraction under the conditions which the container sees the same or substantially the same as that of the plastic from which said container is made;
- 15 (2) a print-receiving area on one major face of layer (1); and
  - or a patterned part of an exposed major face of layer (1) or said print-receiving area (2), said label being regrind compatible with the plastic containers which are to be blow molded around it, said adhesive being characterized by the ability to form a bond with said substrate and said plastic container such that the strength of the substrate-adhesive interface and the plastic container-adhesive interface and the cohesive strength of th adh sive its lf are all great r than th forces r quired for deformation and recovery of th film itself, said label furth r comprising printed indicia on said

print-receiving area (2) of said substrate, or on said substrate at the interface with said adhesive layer (3), or at both locations.

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- 26. A plastic container as defined in Claim 25 wherein said plastic comprises a single or multilayer of polypropylene, polyethylene, polyester or a mixture of any of such plastics.
- 27. A plastic container as defined in Claim 25 wherein said label also includes
- (4) a thin metal coating on at least one of the major faces of substrate layer (1).
- 28. A plastic container as defined in Claim 27 wherein said thin metal coating (4) comprises a vacuum metallized coating of aluminum.
- 29. A plastic container as defined in Claim 25 wherein said layer of substrate (1) comprises a clear or contact clear virgin, recycled or reprocessed polyester.
- 30. A plastic container as defined in Claim 25 wherein said layer of substrate (1) comprises a clear or contact clear virgin, recycled or reprocessed olefin homopolymer or copolymer or a blend thereof.
- 31. A plastic container as defined in Claim 25 further comprising printed indicia on said print-receiving area (2) of said substrate, or on said substrate at the interface with said adhesive layer (3), or at both locations.
- 32. A plastic container as defined in Claim 31 wherein said indicia are reverse printed on said substrate at the interface with said adhesive layer (3) and said adhesive layer overcoats the label completely or in a defined pattern whereby after affixing said label to said plastic container during the blow molding process there will be captured a print d image equivalent to a front label between the label substrate and the container thus prot cting said label from the c ntents of said container wh n spilled, from the

environment and from abrasion when the container is processed, shipped or used.

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- which is clear or translucent and wherein said printing is carried out in three stages so as to produce a first indicia which is reverse printed, a second overprinted indicia which is opaque and a third overprinted indicia which is direct printed whereby after affixing said label to said plastic container during the blow molding process there will be provided in one label the equivalent of a front and back label when the container is empty or partially or completely filled with a clear or translucent liquid.
- 34. A plastic container as defined in Claim 25 wherein said printed indicia is provided with a primer for sealing the printed image and to enhance adhesive bonding if desired.
- 35. A plastic container as defined in Claim 25 wherein said heat activated adhesive layer (3) is a coated layer.
- 36. A plastic container as defined in Claim 25 wherein said heat activated adhesive layer (3) comprises an ethylene/vinyl acetate copolymer.
- 37. A plastic container as defined in Claim 25 wherein the print-receiving face of said substrate (1) has been treated to enhance ink anchorage by application of a primer, by flame-treatment, by corona-treatment or by chemical treatment.

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- 38. A method of producing an in-mold plastic container, said method comprising:
  - (a) providing a polymeric sheet or roll comprising:
- (1) a layer of a transparent, translucent or contact clear substrate formed from a polymer or a mixture of polymers, said layer being surface printable or capable of being rendered surface printable, having a thickness between 0.002 and 0.008 inches, having a specific substantially the same as that of the polymer or mixture of polymers from which it is formed, and having a coefficient of thermal expansion or contraction under the conditions which the container sees the same or substantially the same as that of the plastic from which said container is made;
- (2) a print-receiving area on one major face
  15 of layer (1); and
  - (3) a heat activatable adhesive layer covering all or a patterned part of an exposed major face of layer (1) or said print-receiving area (2), said polymeric sheet being regrind compatible with the plastic containers which are to be blow molded around labels made from it, said adhesive being characterized by the ability to form a bond with said substrate and said plastic container such that the strength of the substrate-adhesive interface and the plastic container-adhesive interface and the cohesive stringth of the adhesive itself are all greater than the forces required for deformation and recovery of the film itself;
  - (b) printing indicia on said print-receiving area(2) of said substrate, or on said substrate at the interface with said adhesive layer (3), or at both locations;
- 30 (c) cutting said sheet into a plurality of individual labels, each label bearing printed indicia;
  - (d) positi ning on of said labels in a mold;

- (e) blow molding a plastic container within said mold such that said label adheres to the outer surface of the35 blow molded container; and
  - (f) removing said container from the mold.
  - 39. A method as defined in Claim 38 wherein said plastic comprises a single or multilayer of polypropylene, polyethylene, polyester or a mixture of any of such plastics.
  - 40. A method as defined in Claim 38 wherein said label also includes
  - (4) a thin metal coating on at least one of the major faces of substrate layer (1).
  - 41. A method as defined in Claim 40 wherein said thin metal coating (4) comprises a vacuum metallized coating of aluminum.
  - 42. A method as defined in Claim 38 wherein said layer of substrate (1) comprises a clear or contact clear oriented virgin, recycled or reprocessed polyester.
  - 43. A method as defined in Claim 38 wherein said layer of substrate (1) comprises a clear or contact clear virgin, recycled or reprocessed olefin homopolymer or copolymer or a blend thereof.
  - 44. A method as defined in Claim 38 further comprising printing indicia on said print-receiving area (2) of said substrate, or on said substrate at the interface with said adhesive layer (3), or at both locations.
  - 45. A method as defined in Claim 44 wherein said indicia are reverse printed on said substrate at the interface with said adhesive layer (3) and said adhesive layer overcoats the sheet completely or in a defined pattern whereby after affixing said label to said plastic container during the blow molding process there will be captured a printed image quivalent to a front label b twe n the label substrate and the container thus protecting said label from the contents of

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said container when spilled, from the environment and from abrasion when the container is processed, shipped or used.

- 46. A method as defined in Claim 45 wherein said printing is carried out in three stages so as to produce a first indicia which is reverse printed, a second overprinted indicia which is opaque and a third overprinted indicia which is direct printed whereby after affixing said label to a clear or translucent plastic container during the blow molding process there will be provided in one label the equivalent of a front and back label when the container is empty or partially or completely filled with a clear or translucent liquid.
- 47. A method as defined in Claim 38 wherein said printed indicia is provided with a primer for sealing the printed image and to enhance adhesive bonding if desired.
- 48. A method as defined in Claim 38 wherein said heat activated adhesive layer (3) is a coated layer.
- 49. A method as defined in Claim 38 wherein said heat activated adhesive layer (3) comprises an ethylene/vinyl acetate copolymer.
- 50. A method as defined in Claim 38 wherein the print-receiving face of said substrate (1) has been treated to enhance ink anchorage by application of a primer, by flame-treatment, by corona-treatment or by chemical treatment.